

AMENDMENTS TO THE CLAIMS

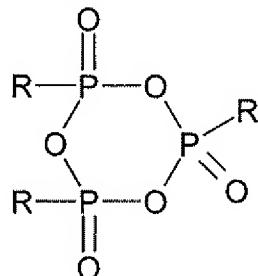
Claims 1-27 (Cancelled)

28. (New) A proton-conducting polymer membrane based on polyazoles which is obtained by a process comprising the steps of
- A) reacting one or more aromatic tetraamino compounds with one or more aromatic carboxylic acids or their esters which contain at least two acid groups per carboxylic acid monomer, or one or more aromatic and/or heteroaromatic diaminocarboxylic acids in the melt at temperatures of up to 350°C,
 - B) dissolving the solid prepolymer obtained in accordance with step A) in an organic phosphonic anhydrides with formation of a solution and/or dispersion,
 - C) heating the solution obtainable in accordance with step B) under inert gas to temperatures of up to 300°C with formation of the dissolved polyazole polymer,
 - D) forming a membrane using the solution of the polyazole polymer in accordance with step C) on a support and
 - E) treating the membrane formed in step D) until it is self-supporting.
29. (New) The membrane as claimed in claim 28 wherein the process comprises the steps of
- A) reacting one or more aromatic tetraamino compounds with one or more aromatic carboxylic acids or their esters which contain at least two acid groups per carboxylic acid monomer, or one or more aromatic and/or heteroaromatic diaminocarboxylic acids in the melt at temperatures of up to 300°C,
 - B) dissolving the solid prepolymer obtained in accordance with step A) in an organic phosphonic anhydrides with formation of a solution and/or dispersion,
 - C) heating the solution obtainable in accordance with step B) under inert gas to temperatures of up to 280°C, with formation of the dissolved polyazole polymer,
 - D) forming a membrane using the solution of the polyazole polymer in accordance with step C) on a support and
 - E) treating the membrane formed in step D) until it is self-supporting.
30. (New) The membrane according to claim 28, wherein said aromatic tetraamino compounds are 3,3',4,4'-tetraaminobiphenyl, 2,3,5,6-tetraaminopyridine, 1,2,4,5-tetraaminobenzene, 3,3',4,4'-tetraaminodiphenyl sulphone, 3,3',4,4'-tetraaminodiphenyl

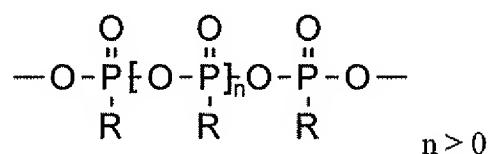
ether, 3,3',4,4'-tetraaminobenzophenone, 3,3',4,4'-tetraaminodiphenylmethane or 3,3',4,4'-tetraaminodiphenyldimethylmethane.

31. (New) The membrane according to claim 28, wherein said aromatic dicarboxylic acids are isophthalic acid, terephthalic acid, phthalic acid, 5-hydroxyisophthalic acid, 4-hydroxyisophthalic acid, 2-hydroxyterephthalic acid, 5-aminoisophthalic acid, 5-N,N-dimethylaminoisophthalic acid, 5-N,N-diethylaminoisophthalic acid, 2,5-dihydroxyterephthalic acid, 2,5-dihydroxyisophthalic acid, 2,3-dihydroxyphthalic acid, 2,4-dihydroxyphthalic acid, 3,4-dihydroxyphthalic acid, 3-fluorophthalic acid, 5-fluoroisophthalic acid, 2-fluoroterephthalic acid, tetrafluorophthalic acid, tetrafluoroisophthalic acid, tetrafluoroterephthalic acid, 1,4-naphthalenedicarboxylic acid, 1,5-naphthalenedicarboxylic acid, 2,6-naphthalenedicarboxylic acid, 2,7-naphthalenedicarboxylic acid, diphenic acid, 1,8-dihydroxynaphthalene-3,6-dicarboxylic acid, diphenyl ether-4,4'-dicarboxylic acid, benzophenone-4,4'-dicarboxylic acid, diphenylsulphone-4,4'-dicarboxylic acid, biphenyl-4,4'-dicarboxylic acid, 4-trifluoromethylphthalic acid, 2,2-bis(4-carboxyphenyl)hexafluoropropane, 4,4'-stilbenedicarboxylic acid, 4-carboxycinnamic acid or their C1-C20 alkyl esters or C5-C12 aryl esters or their acid anhydrides or their acid chlorides.
32. (New) The membrane according to claim 28, wherein said aromatic carboxylic acids are tricarboxylic acids, tetracarboxylic acids or their C1-C20 alkyl esters or C5-C12 aryl esters or their acid anhydrides or their acid chlorides.
33. (New) The membrane according to claim 29, wherein said aromatic carboxylic acids are 1,3,5-benzenetricarboxylic acid (trimesic acid), 1,2,4-benzenetricarboxylic acid (trimellitic acid); (2-carboxyphenyl)iminodiacetic acid, 3,5,3'-biphenyltricarboxylic acid, 3,5,4'-biphenyltricarboxylic acid and/or 2,4,6-pyridinetricarboxylic acid.
34. (New) The membrane according to claim 29, wherein said aromatic carboxylic acids are benzene- 1,2,4,5-tetracarboxylic acid; naphthalene-1,4,5,8-tetracarboxylic acid; 3,5,3',5'-biphenyltetracarboxylic acid; benzophenonetetracarboxylic acid, 3,3',4,4'-biphenyltetracarboxylic acid, 2,2',3,3'-biphenyltetracarboxylic acid, 1,2,5,6-naphthalenetetracarboxylic acid, or 1,4,5,8-naphthalenetetracarboxylic acid.

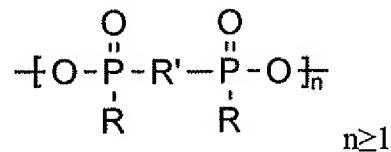
35. (New) The membrane according to claim 32, wherein the content of tricarboxylic acid or tetracarboxylic acids (based on dicarboxylic acid used) is between 0 and 30 mol%.
36. (New) The membrane according to claim 32, wherein the content of tricarboxylic acid or tetracarboxylic acids (based on dicarboxylic acid used) is between 0.5 and 10 mol-%.
37. (New) The membrane according to claim 28, wherein said heteroaromatic carboxylic acids are heteroaromatic dicarboxylic acids, tricarboxylic acids or tetracarboxylic acids, which contain at least one nitrogen, oxygen, sulphur or phosphorus atom in the aromatic group.
38. (New) The membrane according to claim 29, wherein said heteroaromatic carboxylic acids are pyridine-2,5-dicarboxylic acid, pyridine-3,5-dicarboxylic acid, pyridine-2,6-dicarboxylic acid, pyridine-2,4-dicarboxylic acid, 4-phenyl-2,5-pyridinedicarboxylic acid, 3,5-pyrazoledicarboxylic acid, 2,6-pyrimidinedicarboxylic acid, 2,5-pyrazinedicarboxylic acid, 2,4,6-pyridinetricarboxylic acid, benzimidazole-5,6-dicarboxylic acid as well as their C1-C20 alkyl esters or C5-C12 aryl esters or their acid anhydrides or their acid chlorides.
39. (New) The membrane according to claim 28, wherein, in step B), said organic phosphonic anhydrides are of the formula



or linear compounds of the formula

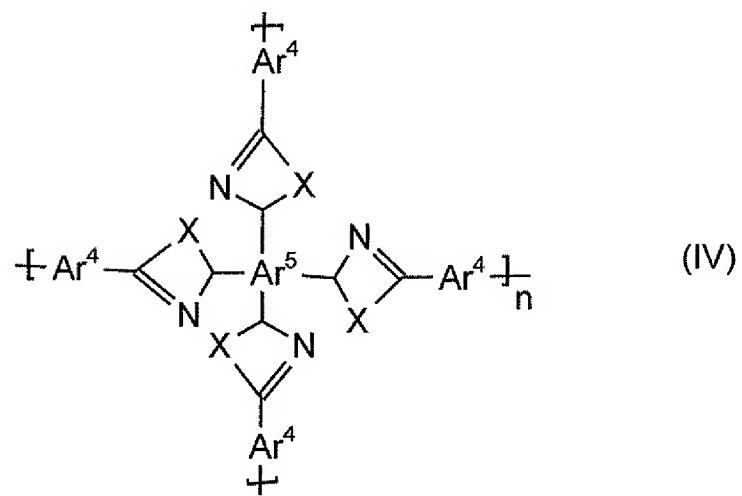
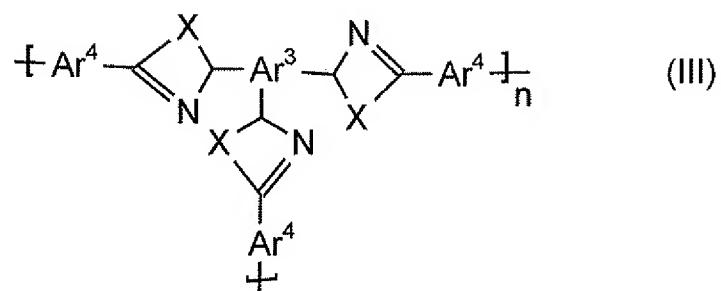
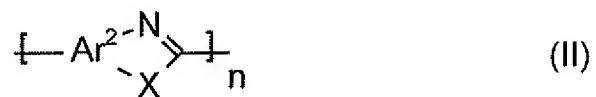
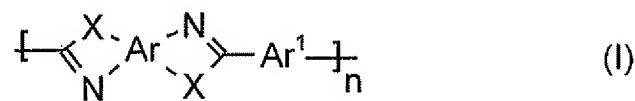


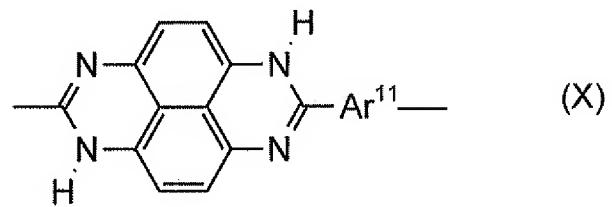
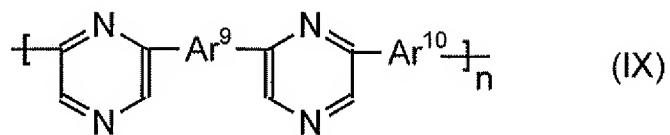
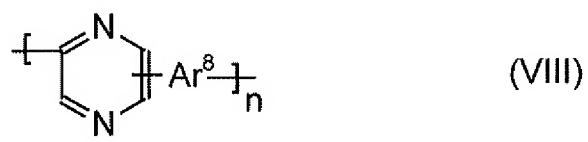
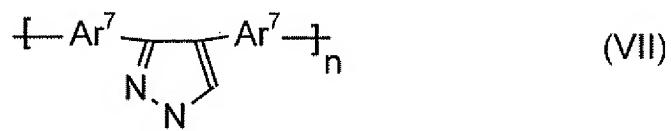
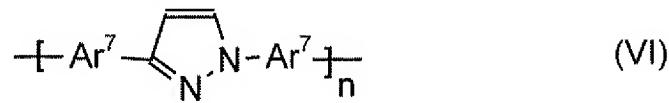
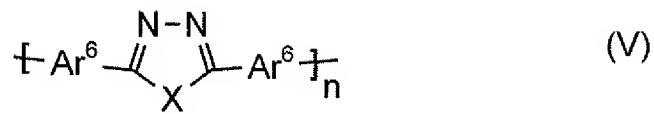
or anhydrides of the multiple organic phosphonic acids of the formula

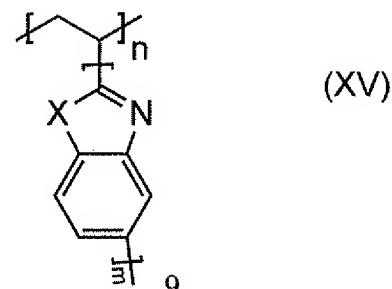
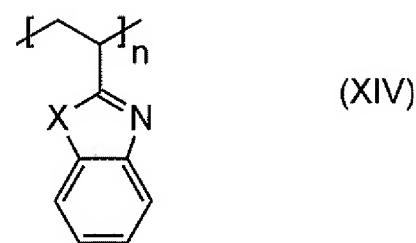
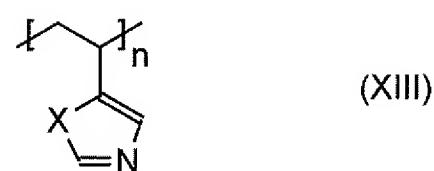
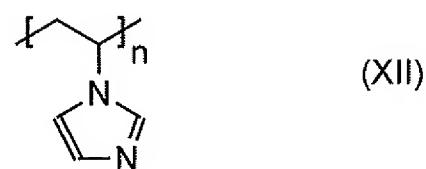
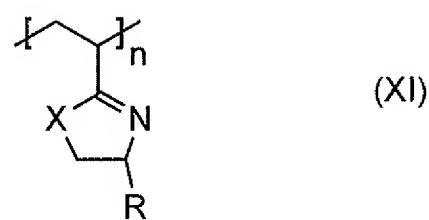


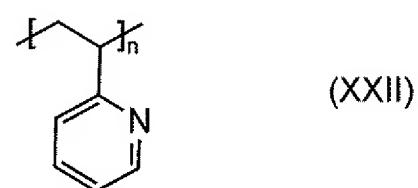
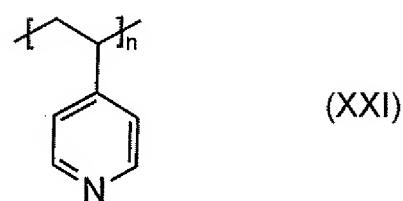
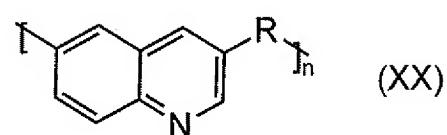
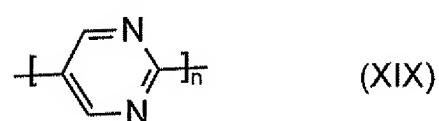
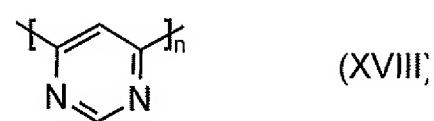
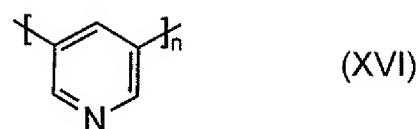
wherein the radicals R and R' are identical or different and represent a C₁-C₂₀ carbon-containing group.

40. (New) The membrane according to claim 28, wherein, in step B), further comprises a polyphosphoric acid having a content of at least 83%, calculated as P₂O₅ (by acidimetry).
41. (New) The membrane according to claim 28, wherein, in step B) further comprises P₂O₅.
42. (New) The membrane according to claim 28, wherein, in step B) or step C), a solution or a dispersion/suspension is produced.
43. (New) The membrane according to claim 28, wherein, in step C), a polymer based on polyazole containing recurring azole units of the formula (I) and/or (II) and/or (III) and/or (IV) and/or (V) and/or (VI) and/or (VII) and/or (VIII) and/or (IX) and/or (X) and/or (XI) and/or (XII) and/or (XIII) and/or (XIV) and/or (XV) and/or (XVI) and/or (XVII) and/or (XVIII) and/or (XIX) and/or (XX) and/or (XXI) and/or (XXII) is formed







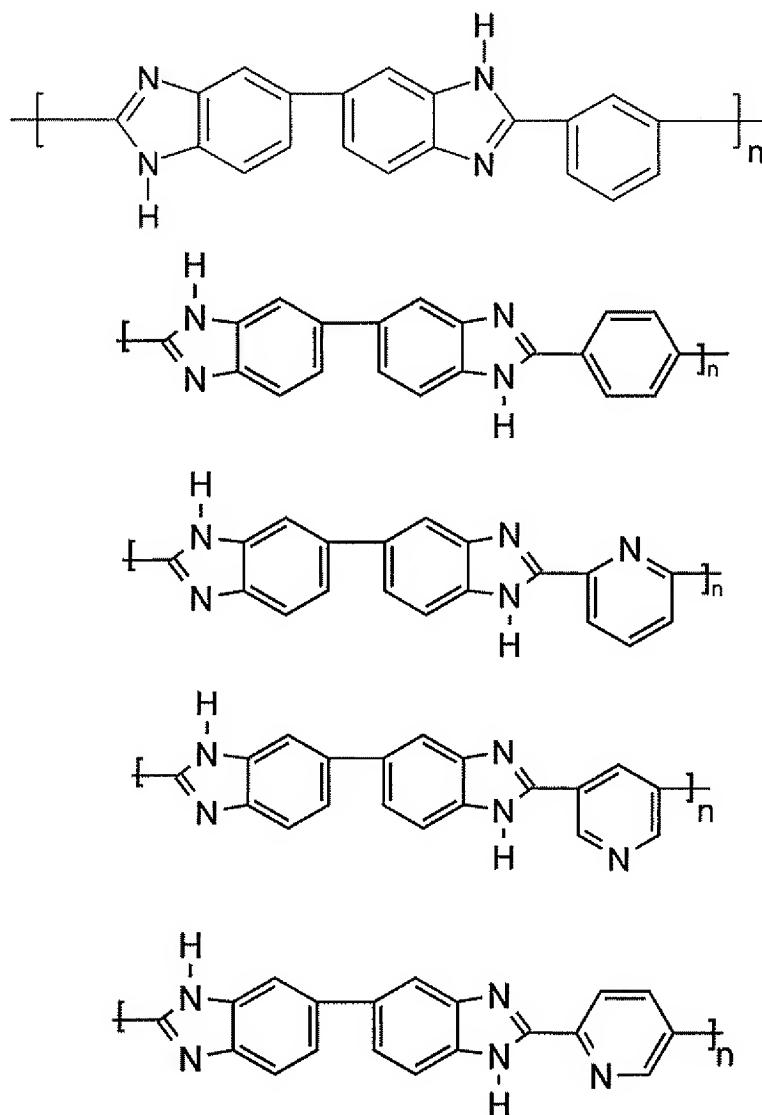


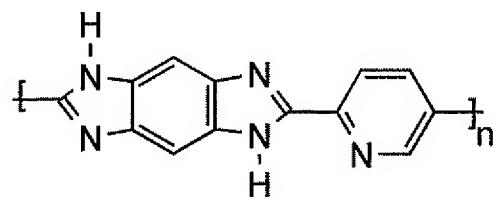
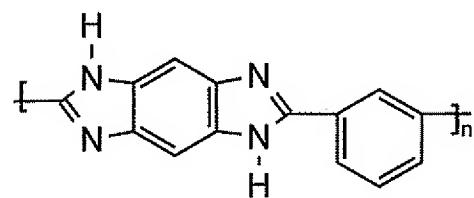
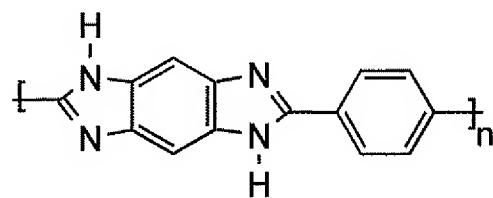
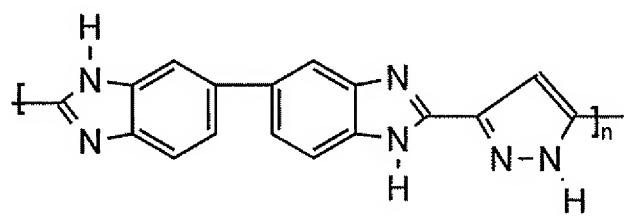
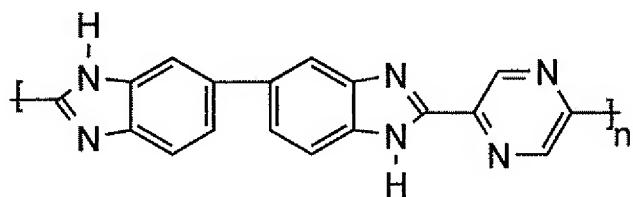
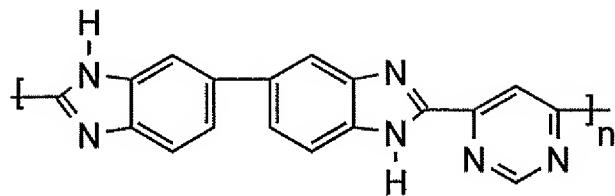
wherein

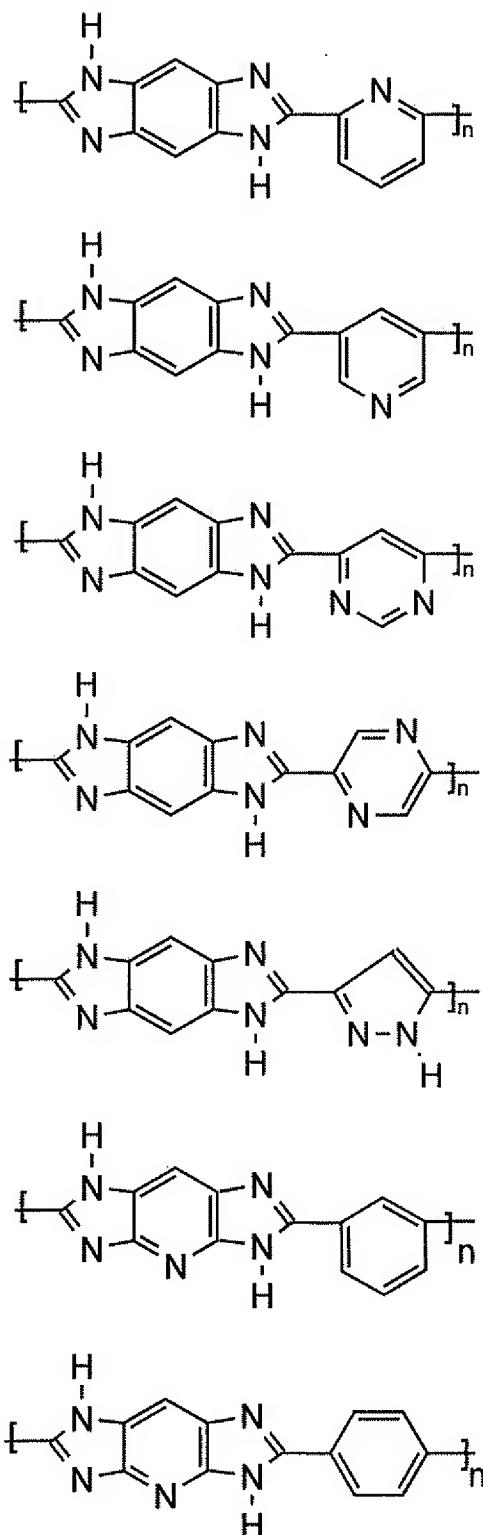
- Ar are identical or different and represent a tetracovalent aromatic or heteroaromatic group which can be mononuclear or polynuclear,
- Ar¹ are identical or different and represent a bicovalent aromatic or heteroaromatic group which can be mononuclear or polynuclear,
- Ar² are identical or different and represent a bicovalent or tricovalent aromatic or heteroaromatic group which can be mononuclear or polynuclear,
- Ar³ are identical or different and represent a tricovalent aromatic or heteroaromatic group which can be mononuclear or polynuclear,
- Ar⁴ are identical or different and represent a tricovalent aromatic or heteroaromatic group which can be mononuclear or polynuclear,
- Ar⁵ are identical or different and represent a tetracovalent aromatic or heteroaromatic group which can be mononuclear or polynuclear,
- Ar⁶ are identical or different and represent a bicovalent aromatic or heteroaromatic group which can be mononuclear or polynuclear,
- Ar⁷ are identical or different and represent a bicovalent aromatic or heteroaromatic group which can be mononuclear or polynuclear,
- Ar⁸ are identical or different and represent a tricovalent aromatic or heteroaromatic group which can be mononuclear or polynuclear,
- Ar⁹ are identical or different and represent a bicovalent or tricovalent or tetracovalent aromatic or heteroaromatic group which can be mononuclear or polynuclear,
- Ar¹⁰ are identical or different and represent a bicovalent or tricovalent aromatic or heteroaromatic group which can be mononuclear or polynuclear,
- Ar¹¹ are identical or different and represent a bicovalent aromatic or heteroaromatic group which can be mononuclear or polynuclear,
- X are identical or different and represent oxygen, sulphur or an amino group which carries a hydrogen atom, a group having 1 - 20 carbon atoms, preferably a branched or unbranched alkyl or alkoxy group, or an aryl group as a further radical,
- R are identical or different and represent hydrogen, an alkyl group and an aromatic group, with the proviso that R in formula (XX) is not hydrogen, and

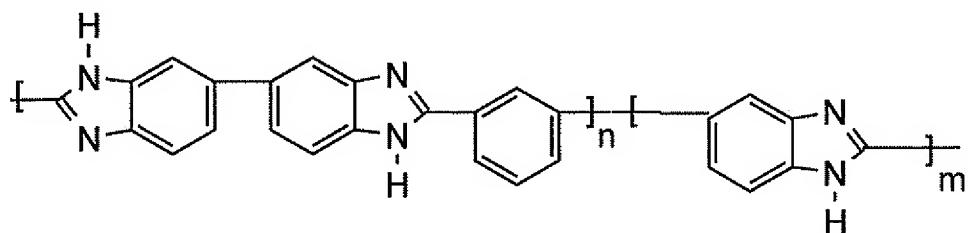
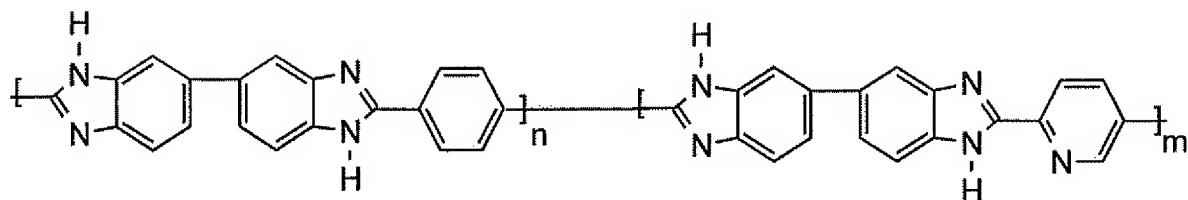
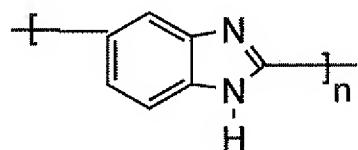
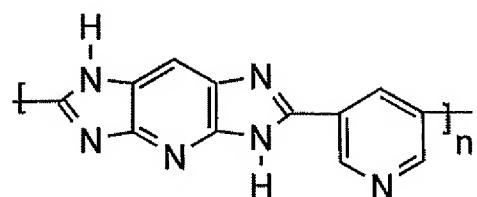
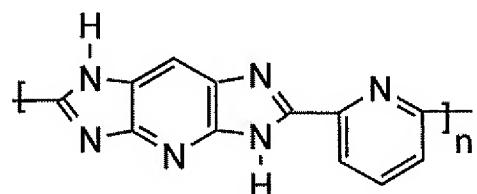
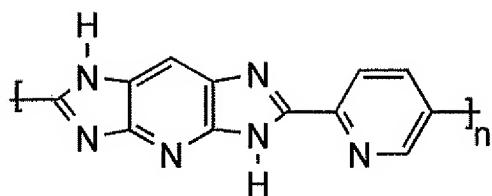
n and m are identical or different and each are an integer greater than or equal to 10.

44. (New) The membrane according to claim 28, wherein, in step C), a polymer selected from the group consisting of polybenzimidazole, poly(pyridines), poly(pyrimidines), polyimidazoles, polybenzothiazoles, polybenzoxazoles, polyoxadiazoles, polyquinoxalines, polythiadiazoles and poly(tetrazapyrenes) is formed.
45. (New) The membrane according to claim 28, wherein, in step C), a polymer containing recurring benzimidazole units of the formula









where n and m are each an integer greater than or equal to 10 is formed.

46. (New) The membrane according to claim 28, wherein, during or after step A), step B), step C), a further polymer is added as blend material.

47. (New) The membrane according to claim 28, wherein, after step C) and before step D), the viscosity is adjusted by addition of phosphoric acid and/or organophosphonic acids.
48. (New) The membrane according to claim 28, wherein the membrane produced in accordance with step E) is treated in the presence of moisture at temperatures and for a period of time until the membrane is self-supporting and can be detached from the support without any damage.
49. (New) The membrane according to claim 28, wherein the treatment of the membrane in step E) is performed at temperatures of more than 0°C and less than 150°C in the presence of moisture or water and/or steam.
50. (New) The membrane according to claim 28, wherein the treatment of the membrane in step E) is performed at temperatures of between room temperature (20°C) and 90°C, in the presence of moisture or water and/or steam and for 1 minute to 200 hours
51. (New) The membrane according to claim 28, wherein the treatment of the membrane in step E) is for 10 seconds to 300 hours.
52. (New) The membrane according to claim 28, wherein, in step D), an electrode is chosen as the support and the treatment in accordance with step E) is such that the membrane formed is no longer self-supporting.
53. (New) The membrane according to claim 28, wherein, in step D), a layer having a thickness of 20 to 4000 µm, preferably between 30 and 3500 µm is produced.
54. (New) The membrane according to claim 28, wherein the membrane formed in step E) has a thickness between 15 and 3000 µm.
55. (New) The membrane according to claim 28, wherein the membrane formed in step E) has a thickness between 20 and 1500 µm.

56. (New) An electrode having a proton-conducting polymer coating based on polyazoles which can be obtained by a process comprising the steps of
- reacting one or more aromatic tetraamino compounds with one or more aromatic carboxylic acids or their esters which contain at least two acid groups per carboxylic acid monomer, or one or more aromatic and/or heteroaromatic diaminocarboxylic acids in the melt at temperatures of up to 350°C,
 - dissolving the solid prepolymer obtained in accordance with step A) in an organic phosphonic anhydrides with formation of a solution and/or dispersion,
 - heating the solution obtainable in accordance with step B) under inert gas to temperatures of up to 300°C with formation of the dissolved polyazole polymer,
 - forming a layer using the solution of the polyazole polymer in accordance with step C) on an electrode and
 - treating the layer formed in step D).
57. (New) The electrode according to claim 56, wherein the coating has a thickness between 2 and 3000 µm.
58. (New) The electrode according to claim 56, wherein the coating has a thickness between 5 and 1500 µm.
59. (New) A membrane electrode unit containing at least one electrode and at least one membrane according to claim 28.
60. (New) The membrane electrode unit containing at least one electrode according to claim 59 and at least one membrane based on polyazoles which is obtained by a process comprising the steps of
- reacting one or more aromatic tetraamino compounds with one or more aromatic carboxylic acids or their esters which contain at least two acid groups per carboxylic acid monomer, or one or more aromatic and/or heteroaromatic diaminocarboxylic acids in the melt at temperatures of up to 350°C,
 - dissolving the solid prepolymer obtained in accordance with step A) in an organic phosphonic anhydrides with formation of a solution and/or dispersion,
 - heating the solution obtainable in accordance with step B) under inert gas to temperatures of up to 300°C with formation of the dissolved polyazole polymer,

- D) forming a membrane using the solution of the polyazole polymer in accordance with step C) on a support and
 - E) treating the membrane formed in step D) until it is self-supporting.
61. (New) A fuel cell containing one or more membrane electrode units according to claim 56.
62. (New) A process to produce a proton-conducting polymer membrane which comprises the steps of
- A) reacting one or more aromatic tetraamino compounds with one or more aromatic carboxylic acids or their esters which contain at least two acid groups per carboxylic acid monomer, or one or more aromatic and/or heteroaromatic diaminocarboxylic acids in the melt at temperatures of up to 350°C,
 - B) dissolving the solid prepolymer obtained in accordance with step A) in an organic phosphonic anhydrides with formation of a solution and/or dispersion,
 - C) heating the solution obtainable in accordance with step B) under inert gas to temperatures of up to 300°C with formation of the dissolved polyazole polymer,
 - D) forming a membrane using the solution of the polyazole polymer in accordance with step C) on a support and
 - E) treating the membrane formed in step D) until it is self-supporting.